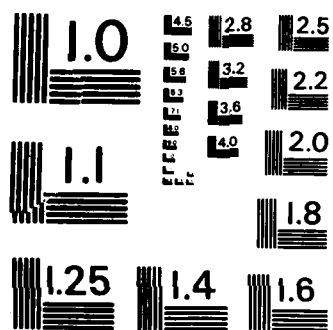


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Transient Excitation of Anharmonic Adspecies by Pulsed Laser Radiation

by

J. T. Lin, Xi-Yi Huang and Thomas F. George

Prepared for Publication

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Departments of Chemistry and Physics
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TRANSIENT EXCITATION OF ANHARMONIC ADSPECIES BY PULSED LASER RADIATION

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ABSTRACT

The excitation of anharmonic adspecies by pulsed laser radiation is investigated theoretically in the transient regimes. New features include the optimum values of pulse duration and detuning and the oscillatory behavior of the time-dependent excitation caused by the anharmonicity.

INTRODUCTION AND ANALYTICAL RESULTS

Laser excitation of adspecies has been extensively studied in the past several years. Such studies, however, have been limited to steady-state excitations and/or adiabatic processes where the laser duration is much longer than the dipole dephasing time. In the present paper, we show an analysis in the transient regimes where new features given by the pulse duration, detuning and anharmonicity are studied both analytically and numerically. The key equations describing the laser excitation of adspecies are given by

$$\frac{da}{dt} = - (i\Delta_{\text{eff}} + \frac{\gamma_1 + \gamma_2}{2}) a - iV(t) , \quad (1)$$

$$\frac{dN}{dt} = iV(t) (a - a^\dagger) - \gamma_1 (N - N_0) , \quad (2)$$

$$\Delta_{\text{eff}} = \Delta - 2\epsilon^* N . \quad (3)$$

Here a and a^\dagger are the dipole operators of the active mode, where $N \equiv \langle a^\dagger a \rangle$, γ_1 and γ_2 are damping factors defined by the inverse of the energy $^1(T_1)$ and the phase (T_2) relaxation time, respectively, and $V(t)$ is the excitation source term proportional to the laser electric field. The initial phonon-bath excitation is given by N_0 , and the nonlinear effects due to the anharmonicity (ϵ^*) are given by Eq. (3), where Δ is the laser detuning for the harmonic case. We shall consider a Gaussian laser profile with pulse duration t_p .

Analytical results are available only for the harmonic cases ($\epsilon^* = 0$), where the excitations at three regimes are found:

- I. Steady-state ($t_p \gg T_{1,2}$): $N_{s.s.} \propto I(t)$ (laser intensity) .
- II. Adiabatic-state ($T_1 > t_p \gg T_2$): $N(t) \propto \phi$ (laser energy) .
- III. Transient-state ($t_p < T_{1,2}$): $N(t) \propto t_p \phi$.

NUMERICAL RESULTS

The role of pulse duration is shown in Fig. 1 for $\gamma_1 = \gamma_2 = 1$ and $\Delta = \epsilon^* = 0$. It is seen that, for a given laser energy, there is an optimum pulse duration and the behavior of the excitations is consistent with the analytical results in the various regimes. The effects of changing the detuning are shown in Fig 2 for $\gamma_1 = \gamma_2 = \epsilon^* = t_p = 1$, and the excitations at various anharmonicities are shown in Fig. 3 for $\Delta = 20$. We note that for a given anharmonicity (detuning), an optimum detuning (anharmonicity) is found accordingly. These features may be realized by the fact that maximal excitation is achieved when the detuning is compensated by the anharmonic excitation term ($2\epsilon^*N$), such that the effective detuning (Δ_{eff}) is suppressed. Furthermore, the significant oscillatory behavior, which is absent in Fig. 1 for the harmonic resonance cases, is found in Fig. 3.

As a concluding remark, we note that the effects caused by an increase in the substrate temperature may be included by introducing a temperature-dependent phonon-bath through N_0 in Eq. (2). Numerical results based upon Eqs. (1)-(3) combined with a heat diffusion equation will be shown elsewhere.

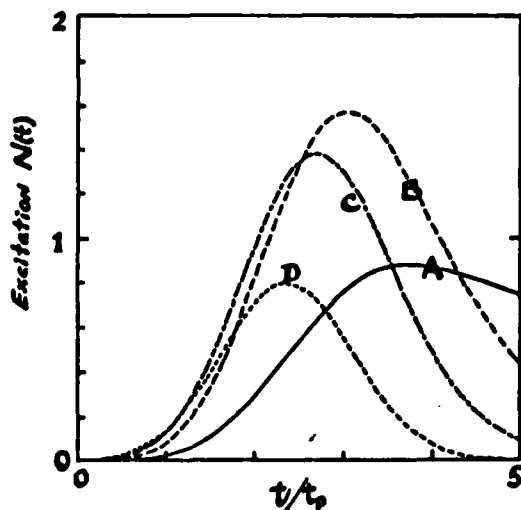


Fig. 1. Harmonic cases with $t_p =$
(A) 0.2, (B) 1, (C) 2 and
(D) 5.

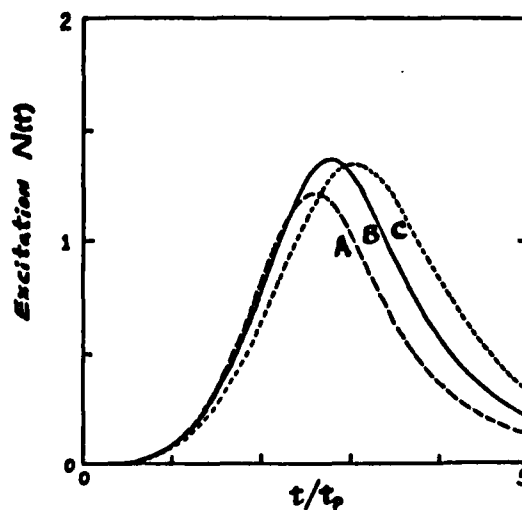


Fig. 2. Anharmonic cases
with $\Delta =$ (A) 0.5, (B) 1.0
and (C) 1.5.

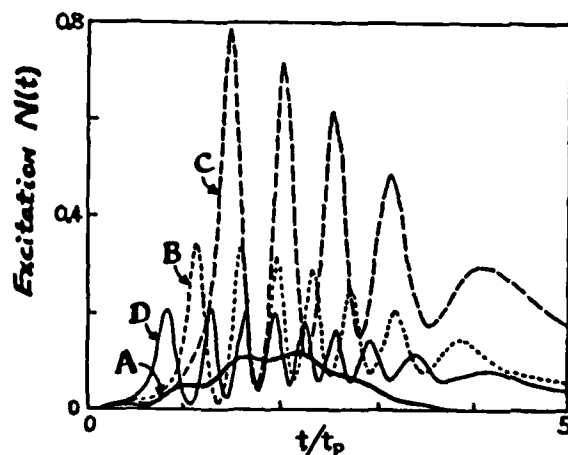


Fig. 3. Oscillatory features for $\epsilon^* =$ (A) 0, (B) 20, (C) 50 and (D) 80.

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